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Molecular Beam Studies of Surface Phonons
and Gas Surface Interactions

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Molecular Beam Studies of Surface Phonons and Gas-Surface Interactions

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The scattering of atomic and molecular beams from well-characterized single crystal surfaces is providing important structural and dynamical information pertaining to clean and adsorbate covered systems. Prime objectives in these studies include the determination of gas-surface interaction potentials, the development of a microscopic understanding of collision induced gas-surface energy transfer, surface crystallography, the elucidation of surface reaction mechanisms, and high resolution ($\sim 1 \text{ cm}^{-1}$) surface vibrational spectroscopy. At the present time especial attention is being given to determining which surface modes actively participate in collision induced energy exchange, the mapping out of surface phonon dispersion relations, and determining under which energy regimes single or multiphonon transitions dominate the interaction.

It is the intent of this progress report to cover some of our recent activities in this field. In particular, the growing importance that angle and velocity resolved inelastic He scattering is playing as a surface spectroscopic tool will be highlighted. These experiments can be visualized as being a two-dimensional (i.e., surface sensitive) analog to inelastic neutron scattering, which has played such a crucial role in elucidating the nature of elementary excitations in bulk crystals.

Experimental data, lattice dynamics calculations, and quantum scattering calculations will be presented which reveal how the surface vibrational properties and geometric structures of ordered Ar, Kr, and Xe rare gas films

evolve, on a layer-by-layer basis, into those of thick crystals. These films form azimuthally aligned but translationally incommensurate structures on the support crystal, Ag(111). The monolayer SP_1 frequencies give valuable information necessary for constructing accurate rare gas-metal physisorption potentials. Deconvoluted inelastic single phonon scattering transition probabilities will also be discussed. Comparison of the experimental phonon dispersion curves with predictions derived from lattice dynamics calculations has allowed us to examine the validity of using gas phase rare gas pair potentials to model condensed phase systems. Examples of the types of data collected are shown in Figures 1 and 2, which respectfully contain time-of-flight spectra and bilayer phonon dispersion curves.

The inelastic scattering experiments are complemented by He diffraction and selective adsorption measurements, which yield information on the nature of He-(rare gas) surface potentials.

Finally, if time permits, a discussion of molecule-surface interactions, adsorbate island characterization, and preliminary results employing a new laser-based particle detector will be briefly presented.

It is with great pleasure that I acknowledge my collaborators and colleagues who made essential contributions to this progress report: K. D. Gibson and C. F. Yu for experimental measurements; K. B. Whaley, E. Pontius and J. C. Light for quantum scattering calculations; and B. M. Hall, D. Mills, and J. E. Black for the theoretical lattice dynamics results. This work was supported, in part, by the Office of Naval Research and the NSF-Materials Research Laboratory Program at The University of Chicago.

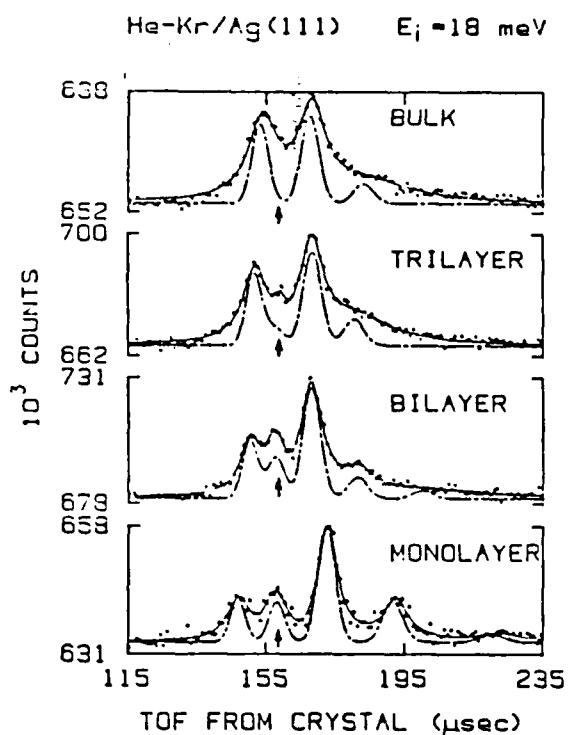


Figure 1. TOF spectra of Kr overlayers physisorbed on Ag(111), $T_s = 25$ K and $E_i = 18$ meV. Crosses are the experimental data, solid lines are least-squares fits to the data, and dashed lines are computer simulations assuming single phonon interactions with delta-function excitation lineshapes. Heights for the simulations were adjusted to be the same as that of the fits. Arrows indicate the position of elastic time-of-flight.

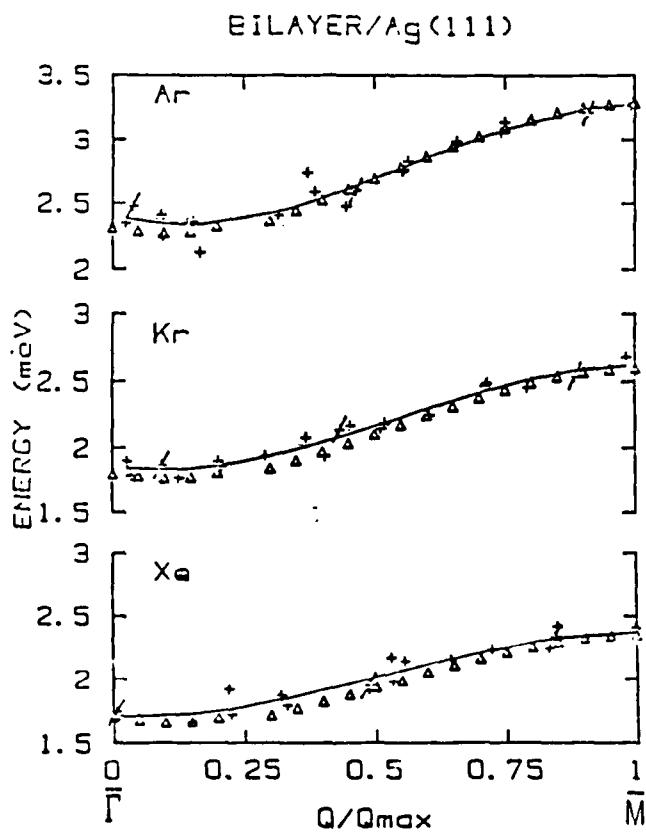


Figure 2. Comparison between the experimental and lattice dynamics results for the bilayer phonon dispersion curves. The crosses are the experimental points, and the slashes through selected crosses are error bars. The solid lines are the best fits to the data. The triangles are the results from lattice dynamics calculations which use as input the experimental lattice constants and realistic two-body Ar-Ar, Kr-Kr, and Xe-Xe pair potentials.